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Publication date:
2015

Document Version
Peer reviewed version

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In situ environmental transmission electron microscopy (ETEM) is of great importance to provide direct experimental evidence of structural dynamics of nanomaterials in a reactive gas at elevated temperature.[1,2] Here, the catalytic nucleation of carbon nanostructures from nanoparticles are recorded with aberration-corrected high-resolution TEM studies. Figure 1. shows the surface-bounded graphene on a certain facet of the particle stacks layer by layer, meanwhile, the disordered region inside the particle expands deeply along the interface of particle and support, suggesting a bulk diffusion path of carbon. However, the appearance of the gas-absorbed interface on the other facet of the particle allows assembling a graphene layer on its partial surface via surface diffusion. The evolution of the particle morphology then results in an increased number of layers and a tube-like structure. Overall, this work provides an improved understanding of nucleation mechanism of graphene and carbon nanotubes.

(1) Hansen, T. W.; Wagner, J. B. *ACS Catal.* **2014**, *4*, 1673.
(2) He, M.; Kauppinen, E. I. et al. *Sci. Rep.* **2013**, *3*.

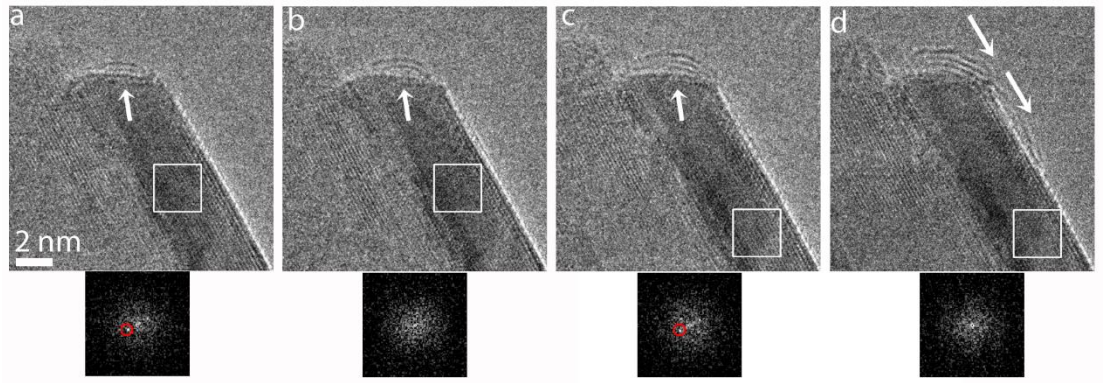

Figure 1. A series of high-resolution TEM images showing the nucleation processes of multilayer graphene. (a-c) A stack of graphene on one facet of the particle. (d) A graphene layer assembles on the other facet of the particle. Fast fourier transforms (FFTs) from the squares showing the expanded disordered region inside the particle.